

Elucidating Mechanisms of Silica Scaling in Membrane Distillation: Effects of Membrane Surface Wettability

Supporting Information

Environmental Science: Water Research & Technology

Yiming Yin¹, Wei Wang², Arun K. Kota², Song Zhao³, and Tiezheng Tong^{1*}

¹ *Department of Civil and Environmental Engineering, Colorado State University, Fort Collins, Colorado 80523, United States*

² *Department of Mechanical Engineering, Colorado State University, Fort Collins, Colorado 80523, United States*

³ *School of Chemical Engineering and Technology, Tianjin Key Laboratory of Membrane Science and Desalination Technology, Tianjin University, Tianjin 300072, P. R. China*

* Corresponding author: email: tiezheng.tong@colostate.edu; Tel. +1 (970) 491-1913

Number of Pages: 12

Number of Figures: 9

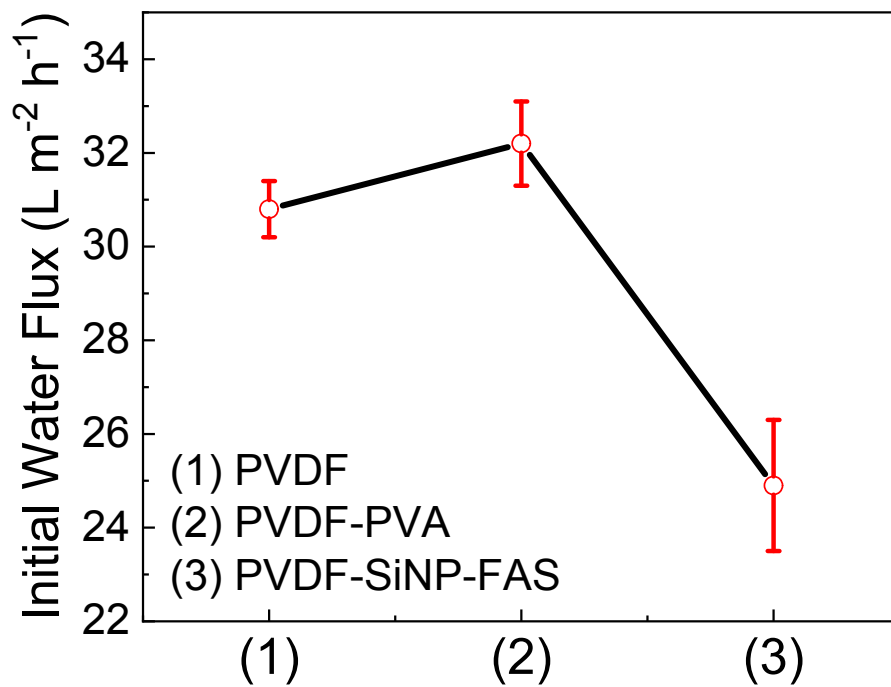
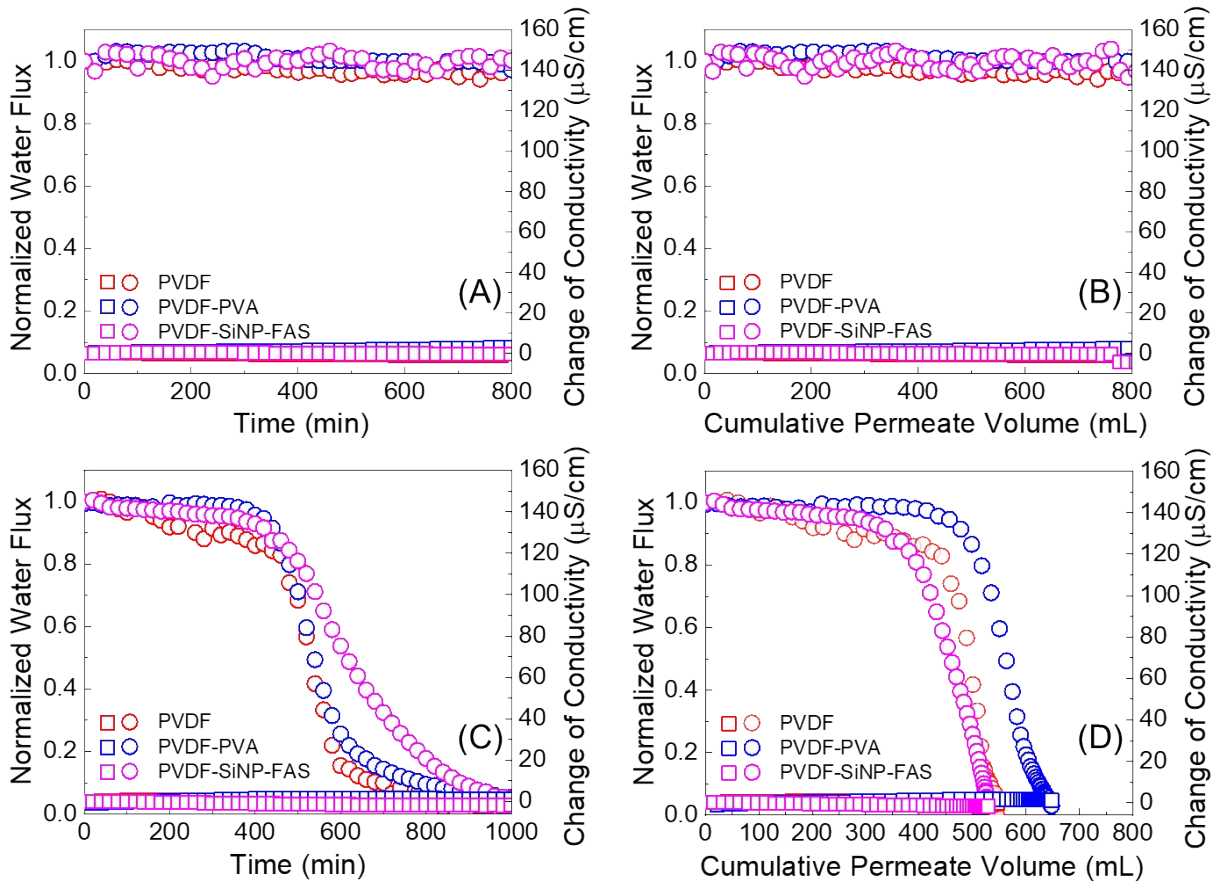


Figure S1. The initial water vapor fluxes of the PVDF, PVDF-PVA and PVDF-SiNP-FAS membranes during direct contact membrane distillation (DCMD) experiments. The error bars were calculated from four independent experiments.



Figures S2. Normalized water flux and change of distillate conductivity of PVDF, PVDF-PVA and PVDF-SiNP-FAS membranes during the DCMD scaling experiment. The feedwater contained 50 mM NaCl, 1 mM NaHCO_3 , as well as 1.5 mM $\text{Na}_2\text{SiO}_3 \cdot 5\text{H}_2\text{O}$ (A, B) or 6 mM $\text{Na}_2\text{SiO}_3 \cdot 5\text{H}_2\text{O}$ (C, D) at pH of 6.50 ± 0.05 . The crossflow velocities in the feed and distillate streams were 9.6 cm/s and 6.4 cm/s, respectively. The temperatures for feed and distillate streams were maintained at 60°C and 20°C , respectively. The initial volume of feed solution was 1500 ml. The initial water vapor fluxes were $30.7 \pm 0.2 \text{ L m}^{-2} \text{ h}^{-1}$, $33.2 \pm 0.25 \text{ L m}^{-2} \text{ h}^{-1}$, and $24.0 \pm 0.7 \text{ L m}^{-2} \text{ h}^{-1}$ for PVDF, PVDF-PVA and PVDF-SiNP-FAS membranes, respectively. These results serve as independent replicate results of what are presented in Figure 2 of the main text.

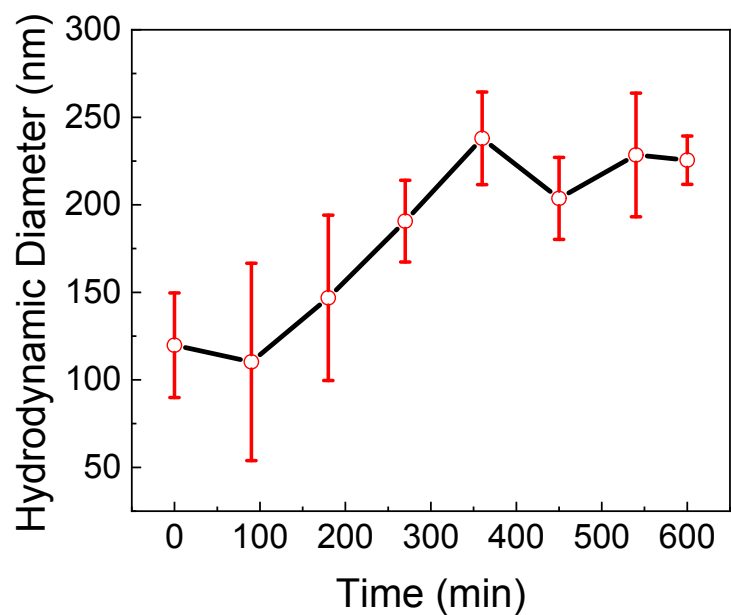


Figure S3. The hydrodynamic diameters of particles formed during DCMD of feedwater containing 6 mM of $\text{Na}_2\text{SiO}_3 \cdot 5\text{H}_2\text{O}$. The error bars were calculated from five independent measurements.

Calculation of concentration polarization of Na₂SiO₃ for different membranes

The concentration of Na₂SiO₃ on the membrane surface is higher than that of the bulk solution due to concentration polarization (CP). Since the extent of CP relates to the water flux across the membrane, we evaluated the CP modulus of Na₂SiO₃ for each membrane used in our study. According to the film model [1], the scalant concentration at the membrane surface (C_m) can be estimated using the following equations:

$$\frac{C_m}{C_b} = \exp\left(\frac{J_w}{k}\right) \quad (1)$$

$$k = \frac{Sh \cdot D}{d_h} \quad (2)$$

where C_b is the scalant concentration in the bulk solution, J_w is the measured water vapor flux, k is the mass transfer coefficient, Sh is the Sherwood number, D is the solution diffusion coefficient, and d_h is the hydraulic diameter.

However, we could not find the accurate D value of Na₂SiO₃ at 60 °C. In the reference of Rebreau et al. [2], the D values of dissolved silica in the temperature range of 2-30 °C were reported. Assuming a linear relation between temperature and diffusion coefficient, we obtained the D value of 1.9×10^{-9} m²/s for dissolved silica. As a result, the CP moduli of Na₂SiO₃ were calculated as 1.7, 1.7, 1.5 for pristine PVDF membrane, PVDF-PVA membrane, and PVDF-SiNP-FAS membrane, respectively.

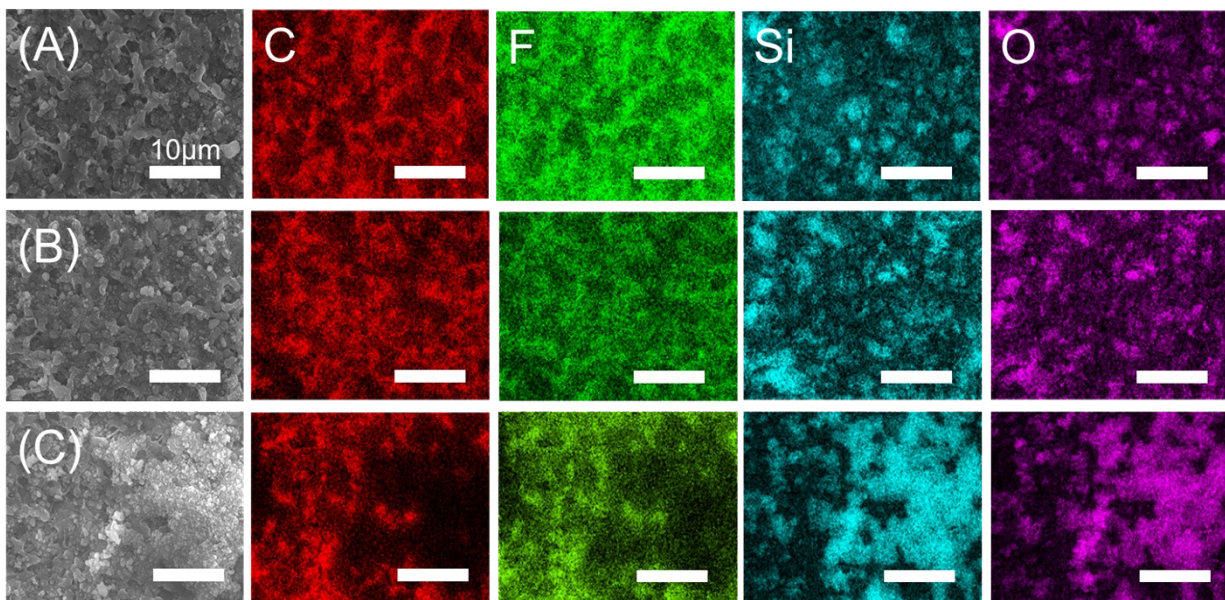


Figure S4. SEM-EDS elemental analysis of the PVDF (A), PVDF-PVA (B) and PVDF-SiNP-FAS (C) membranes (top-view) after DCMD silica scaling experiments. The concentration of silica in the feed solution was 6 mM.

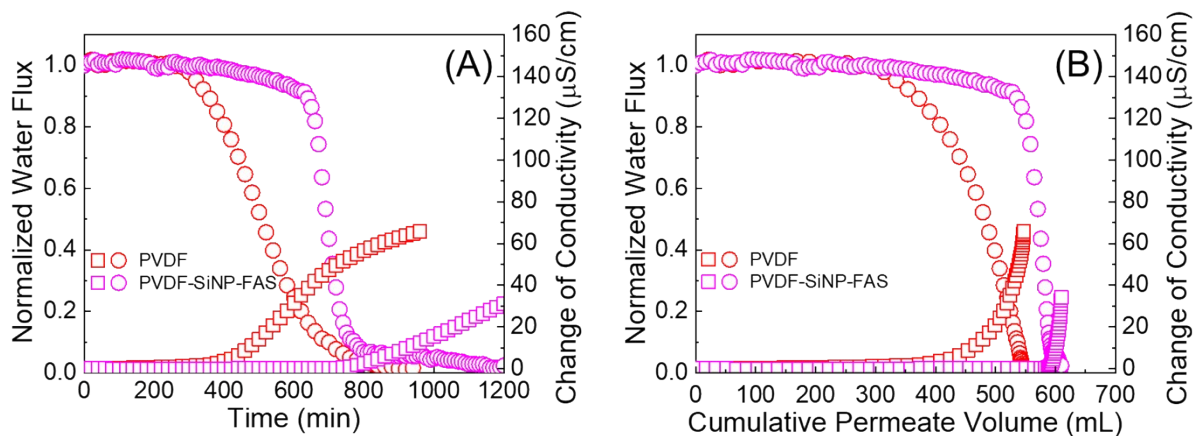


Figure S5. Normalized water flux and change of distillate conductivity of PVDF (red) and PVDF-SiNP-FAS (purple) membranes during DCMD experiments with gypsum scaling. The feed solutions contained 20 mM CaCl_2 , 20 mM Na_2SO_4 , and 50mM NaCl . The crossflow velocities in the feeding and permeating streams were 9.6 cm/s and 6.4 cm/s, respectively. The temperatures for feed and distillate streams were maintained at 60°C and 20°C, respectively. The initial volume of feed solution was 1500 ml. The initial water vapor fluxes were 30.9 $\text{L m}^{-2} \text{h}^{-1}$ and 25.5 $\text{L m}^{-2} \text{h}^{-1}$ for PVDF and PVDF-SiNP-FAS membranes, respectively.

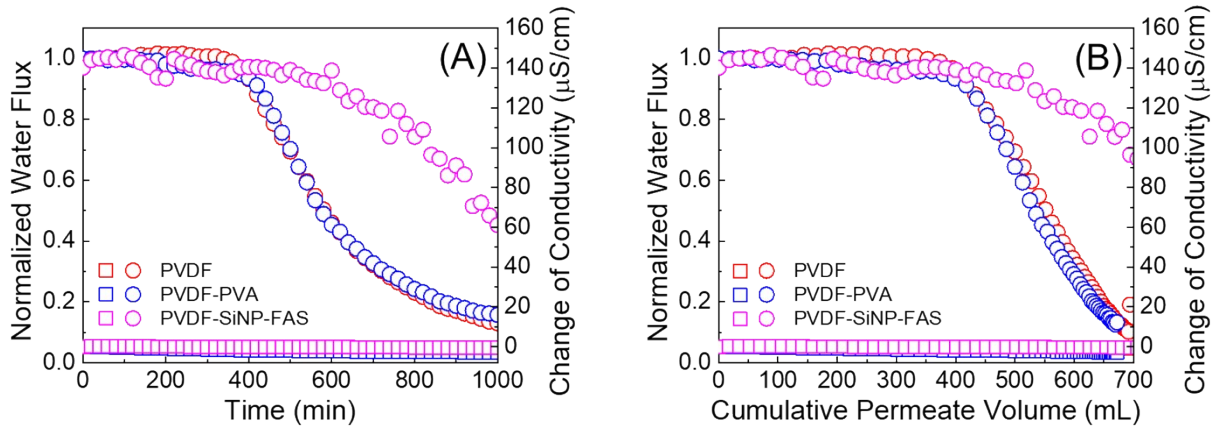


Figure S6. Normalized water flux and change of distillate conductivity of PVDF, PVDF-PVA and PVDF-SiNP-FAS membranes during DCMD experiments with colloidal silica fouling. The feed solutions contained 50 mM NaCl, 1 mM NaHCO₃, and 360 mg/L silica colloidal particles at pH of 6.50 ± 0.05. The crossflow velocities in the feeding and permeating streams were 9.6 cm/s and 6.4 cm/s, respectively. The temperatures for feed and distillate stream were maintained at 60°C and 20°C, respectively. The initial volume of feed solution was 1500 ml. The initial water fluxes were 30.3 L m⁻² h⁻¹, 30.4 L m⁻² h⁻¹, and 25.9 L m⁻² h⁻¹ for PVDF, PVDF-PVA and PVDF-SiNP-FAS membranes, respectively. These results serve as independent replicate results of what are presented in Figure 6 of the main text.

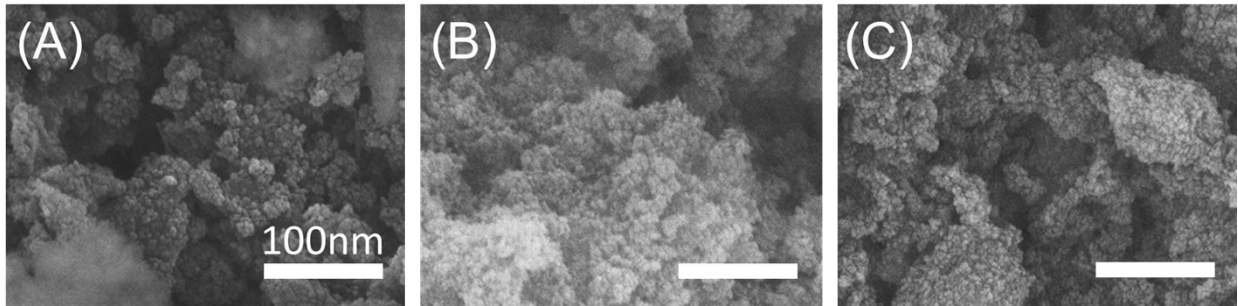


Figure S7. SEM micrographs of PVDF (A), PVDF-PVA (B) and PVDF-SiNP-FAS (D) membranes after DCMD experiments with colloidal silica fouling. The concentration of silica particles was 6 mM. Note that in contrast to silica scaling (Figure 4D-F), no compact and gel-like scale layer was observed.

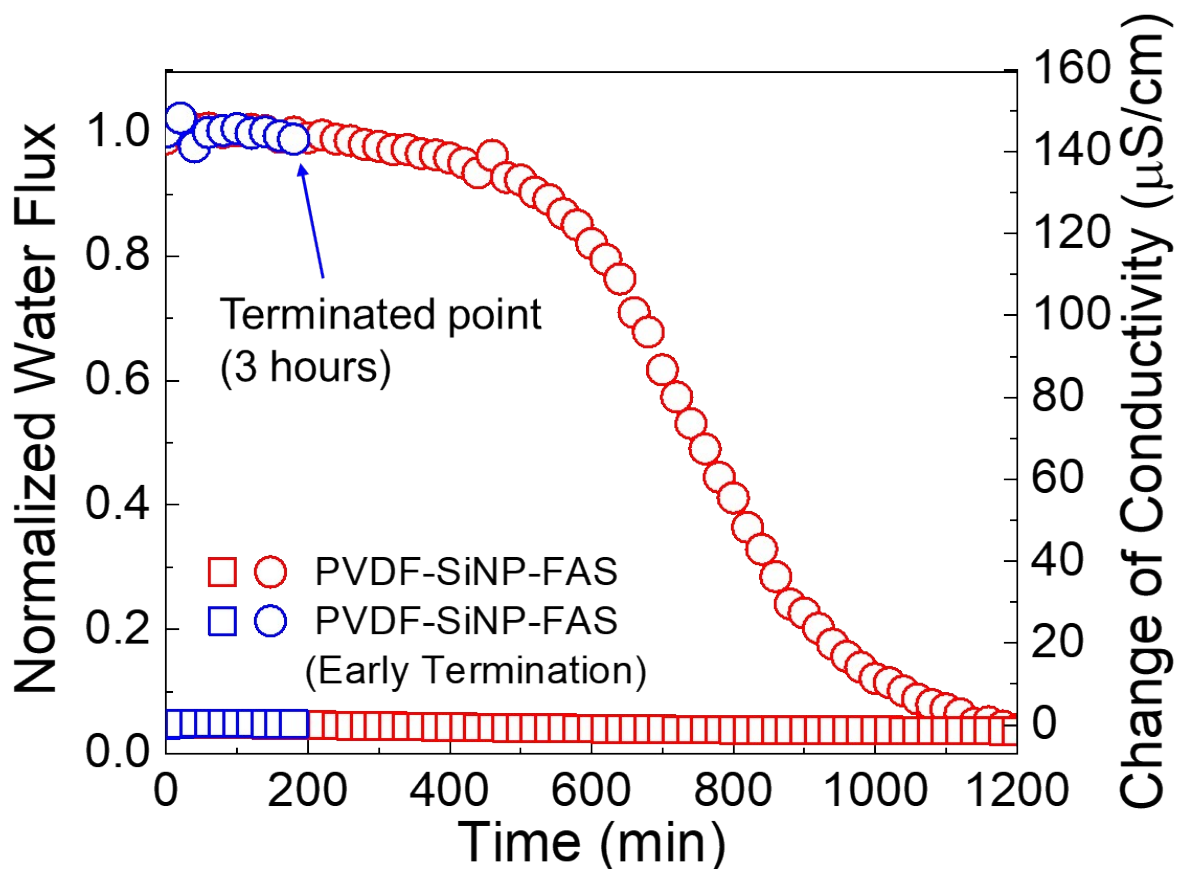


Figure S8. Normalized water flux and change of distillate conductivity for PVDF-SiNP-FAS membranes during DCMD experiments with silica fouling. The experiment was terminated at 3 hours (blue) for the analysis of membrane surface wettability after short-term scaling tests. A full scaling experiment (red), which was performed for > 1000 minutes, is presented for comparison. The feed solutions contained 50 mM NaCl, 1 mM NaHCO₃, and 6 mM Na₂SiO₃·5H₂O at pH of 6.50 ± 0.05. The crossflow velocities in the feeding and permeating streams were 9.6 cm/s and 6.4 cm/s, respectively. The temperatures for feed and distillate stream were maintained at 60°C and 20°C, respectively. The initial volume of feed solution was 1500 ml.

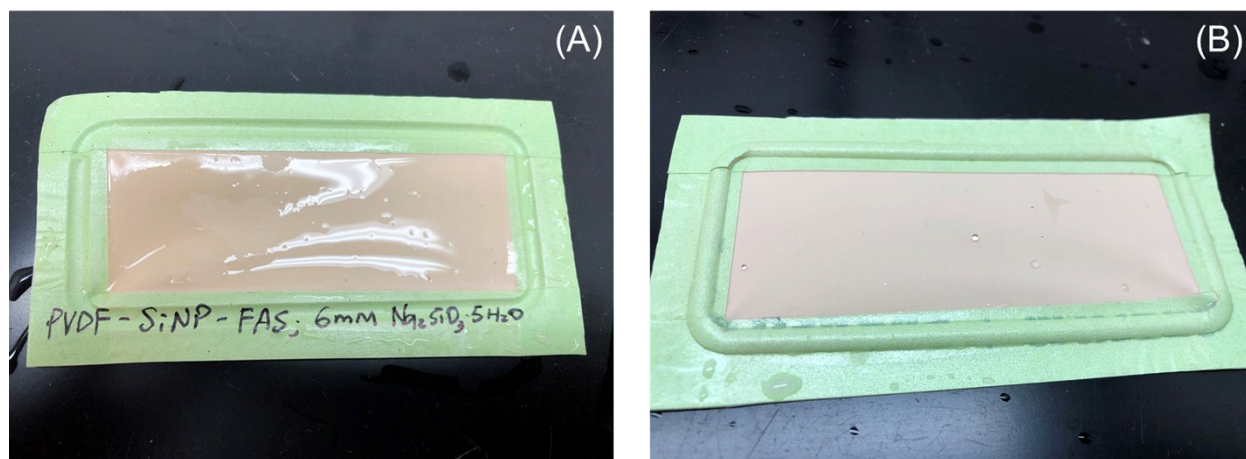


Figure S9. Photographs of PVDF-SiNP-FAS membrane after 3 hours of DCMD treatment of feed solution containing 6 mM $\text{Na}_2\text{SiO}_3 \cdot 5\text{H}_2\text{O}$ (as shown in Figure S8). (A) The membrane side facing the feed stream; and (B) the membrane side facing the distillate stream.

References

- [1] W.L. Qin, J.H. Zhang, Z.L. Xie, D. Ng, Y. Ye, S.R. Gray, M. Xie, Synergistic effect of combined colloidal and organic fouling in membrane distillation: Measurements and mechanisms, *Environ Sci-Wat Res*, 3 (2017) 119-127.
- [2] L. Rebreau, J.P. Vanderborght, L. Chou, The diffusion coefficient of dissolved silica revisited, *Mar Chem*, 112 (2008) 230-233.